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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Group Art Unit: 1209  
Application of: Garcia et al.  
Serial No.: 08/479,077  
Filed: June 6, 1995  
For: SQUARYLIUM COMPOUNDS, AND  
PROCESSES AND INTERMEDIATES FOR THE  
SYNTHESIS OF THESE COMPOUNDS  
Examiner: Cebulak, M.

Cambridge, Massachusetts  
June 13, 1997

APPEAL BRIEF

Assistant Commissioner for Patents  
Washington DC 20231

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## **AUTHORITY RELIED UPON BY APPLICANTS**

*In re Marosi, Stabenow, and Schwarzmann*, 218 USPQ 289 (CAFC 1983)

Sir:

This is an appeal from the final rejection of all claims of the above application as set forth in the Office Action (Paper No. 7) mailed January 7, 1997.

## **REAL PARTY IN INTEREST**

The real party in interest in this appeal is Polaroid Corporation, a corporation organized and existing under the laws of the State of Delaware, of 549 Technology Square, Cambridge, MA 02139-3589.

## **RELATED APPEALS AND INTERFERENCES**

Application Serial No. 08/052,212, from which the present application is divided, is currently on appeal; the rejections being considered in this appeal include, *inter alia*, a 35 USC 103 rejection based, in part, upon U.S. Patent 4,508,811 to Gravesteijn, the same reference being applied in the present case. Although the rejections under appeal in Application Serial No. 08/052,212 are not very similar to those in the present appeal, in the interests of efficiency, the Office may wish to have the same panel consider both appeals. There are no related interferences.

## **STATUS OF CLAIMS**

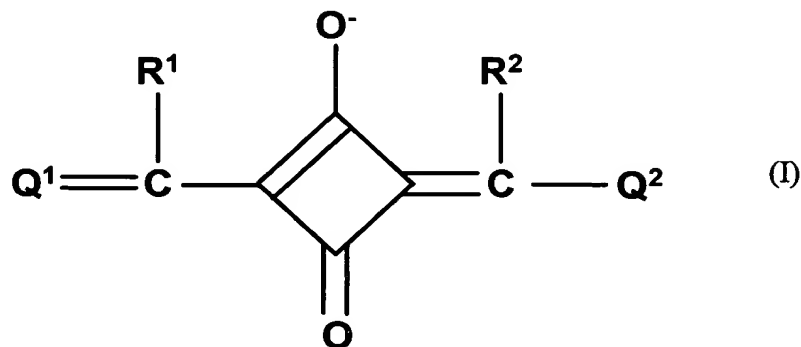
Claims 15-24 are pending in this application, claims 14 and 25-44 having been cancelled; claims 1-13 were cancelled at the time of filing this divisional application. Claims 15-24 stand finally rejected; no claim is objected to.

## **STATUS OF AMENDMENTS**

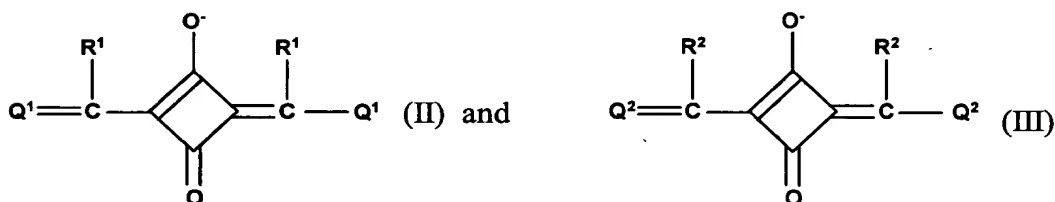
No Amendment After Final Rejection has been filed, so all Amendments have been entered.

## SUMMARY OF INVENTION

This invention relates to a squarylium compound of the formula:



wherein  $Q^1$  and  $Q^2$  are each independently a pyrylium, thiopyrylium, selenopyrylium, benzpyrylium, benzthiopyrylium or benzselenopyrylium nucleus, and  $R^1$  and  $R^2$  are each independently a hydrogen atom or an aliphatic or cycloaliphatic group, the  $Q^1CR^1$  grouping being different from the  $Q^2CR^2$  grouping. In a preferred form of the invention, claimed in claim 16, the compound of Formula I is essentially free from squarylium compounds of the formulae:



It is known that compounds in which two heterocyclic nuclei are linked by a pentamethine chain, the three central carbon atoms of which form part of a squarate ring, are useful as dyes, especially near infra-red dyes. (The term "near infra-red" is used herein to mean electromagnetic radiation having a wavelength of about 700 to about 1200 nm.) For example, Japanese Patent Application No. 103,604/82 discloses a broad class of bis-heterocyclic pentamethine dyes in which the central three carbon atoms of the pentamethine chain form part of a squarylium or croconylium ring. The heterocyclic nuclei can be pyrylium, thiopyrylium, selenopyrylium, benzpyrylium, benzthiopyrylium, benzselenopyrylium, naphthopyrylium,

naphthothiopyrylium or naphthoselenopyrylium nuclei, which can be substituted with alkyl, alkoxy, aryl or styryl groups.

Most of the known pentamethine squarate dyes are symmetrical, that is to say the two heterocyclic nuclei are the same. Such symmetrical dyes are typically prepared by condensing two moles of the appropriate alkyl-substituted heterocyclic compound (usually, a salt) with squaric acid in the presence of a base.

In certain applications of pentamethine squarate dyes, it may be advantageous to use a dye which is asymmetric, i.e., which contains two different heterocyclic groupings. For example, some symmetrical near infra-red pentamethine squarate dyes have significant absorption in the visible region, and this visible absorption restricts the utility of the dyes in certain applications, for example thermal imaging media. In particular, if the symmetrical dye absorbs strongly in one part of the visible spectrum but not in another, it will tend to introduce color distortion into any image created using the symmetrical dye. Although asymmetrical analogues of these infra-red pentamethine squarate dyes may have some visible absorption, this visible absorption tends to take the form of several separate small peaks, and is thus more spread out over a wide range of wavelengths than in the symmetrical dyes. Such absorption over a range of wavelengths tends to produce lower peak absorption and less color distortion (because the dye tends to produce a gray tint) than that produced by the symmetrical dyes, and thus the asymmetric dyes may advantageously be used in applications where the visible absorption of the symmetric dyes causes problems.

Moreover, there are a number of applications where infra-red dyes are needed which absorb at specific wavelengths. For example U.S. Patents Nos. 4,602,263 and 4,826,976 both describe thermal imaging systems for optical recording and particularly for forming color images. These patents describe a preferred form of thermal imaging medium for forming multicolor images; in this preferred imaging medium, three separate color-forming layers, capable of forming yellow, cyan and magenta dyes respectively, are superposed on top of one another. Each of the three color-forming layers has an associated infra-red absorber, these absorbers absorbing at

differing wavelengths, for example 760, 820 and 880 nm. This medium is imagewise exposed simultaneously to three lasers having wavelengths of 760, 820 and 880 nm. The resultant imagewise heating of the color-forming layers causes the leuco dyes to undergo color changes in the exposed areas, thus producing a multicolored image, which needs no development. If the choice of infra-red dyes is restricted to symmetrical compounds, it may be difficult to find a dye which absorbs at the precise wavelength required, and which meets the other requirements, such as storage stability and miscibility in polymers, for use in such media. Asymmetric dyes, which allow the two groups linked to the squarylium nucleus to be varied independently, provide an extra degree of freedom, which renders it easier to find a dye which absorbs at the desired wavelength and meets the other requirements for use in such media.

However, despite the potential advantages of asymmetric pentamethine squarate dyes, little research has been conducted on such dyes because of the difficulties involved in their synthesis. Although it is in theory possible to modify the conventional alkyl-substituted heterocyclic compound/squaric acid condensation reaction to produce asymmetric pentamethine dyes by including two different heterocyclic compounds in the reaction mixture, such a modified process inevitably produces three different products (two symmetrical dyes and the desired asymmetric dye), thus wasting at least half the starting materials (and possibly more if one heterocyclic compound is significantly more reactive than the other). Given that the costs of some symmetric pentamethine squarate dyes are high, such materials should be used judiciously and their loss minimized where possible.

Furthermore, separation of the tertiary product mixture produced is difficult, especially since, in many cases of practical importance, the two heterocyclic compounds used are chemically similar. For example, if one attempts to produce the dye of Formula A shown in Figure 1 of this application in which  $R^1$  and  $R^2$  are each a hydrogen atom (this dye contains one pyrylium nucleus and one selenopyrylium nucleus) simply by condensing a mixture of the two corresponding salts with squaric acid, it is extremely difficult to separate the desired asymmetric salt from the two,

even on a laboratory scale, and conducting this separation on a commercial scale would be a practical impossibility. In some applications of infra-red dyes, the presence of even minor amounts of symmetric by-products in the desired asymmetric dye may cause significant problems. For example, as already noted, in the thermal imaging media described in the aforementioned U.S. Patents Nos. 4,602,263 and 4,826,976, three separate imaging layers are present having infra-red absorbers with absorptions at 760, 820 and 880 nm. Conveniently, two of these three absorbers are Dye A shown in Figure 1, in which R<sup>1</sup> and R<sup>2</sup> are each a hydrogen atom, and the corresponding bis-selenopyrylium dye. However, if Dye A is contaminated with even a small proportion of the corresponding bis-selenopyrylium dye, serious problems may result in such a medium, in that the bis-selenopyrylium impurity in the layer containing Dye A will absorb the "wrong" radiation, which may lead to unwanted exposure of parts of the layer containing Dye A and a reduction in sensitivity of the medium because the bis-selenopyrylium impurity will absorb a large part of the radiation intended to cause color change in a different color-forming layer.

There is thus a need to prepare asymmetric pentamethine squarate dyes without separation of mixtures of asymmetric and symmetric products. The present inventors provide such a preparation by the process shown, for example, in Figures 1 and 2 of the application, in which a squaric acid derivative is condensed with one mole of a pyrylium (or similar) salt to form a "monocondensed intermediate" of Formula E shown in Figure 1, followed by condensation of this monocondensed intermediate with one mole of a different pyrylium (or similar) salt to form the asymmetric final product. This process has been acknowledged to be new and non-obvious, as shown by the issue of U.S. Patent No. 5,231,190, issued on the grandparent Application Serial No. 07/696,222. The process produces the asymmetric dye essentially free from contamination by either of the corresponding symmetric dyes.

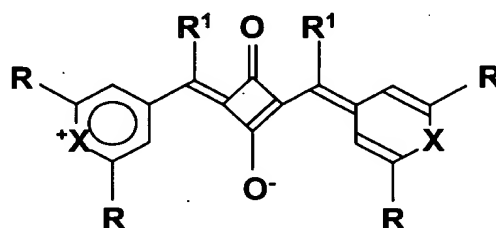
## REFERENCES APPLIED BY EXAMINER

U.S. Patent No. 4,508,811 (Gravesteijn et al.)

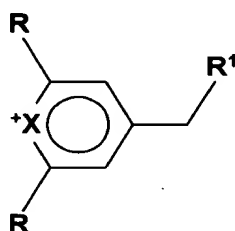
U.S. Patent No. 5,079,127 (Katagiri)

## EXPLANATION OF THE REFERENCES

Gravesteijn describes an optical recording element (i.e., a compact disc) which uses in its dye layer a squarylium dye of formula:



in which R is an alkyl group having at least three carbon atoms, and R<sub>1</sub> is a hydrogen atom or methyl. This dye is produced by reacting two moles of the corresponding 4-alkylpyrylium salt of the formula:

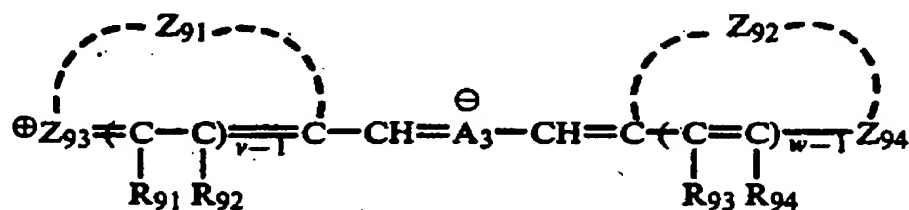


with squaric acid.

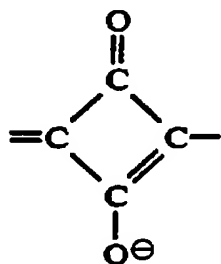
In the dye of the first formula given above, "X is an oxygen atom or a sulfur atom" (see column 2, line 26 and claim 1 of Gravesteijn). The patent never states whether the two atoms X must be the same or can be different. However, the only synthetic Examples, Examples 1 and 2, show preparations of dyes in which both atoms X are sulfur (Example 1) or oxygen (Example 2); there is no description of a dye in which one atom X is sulfur and the other is oxygen. Furthermore, the detailed description of the synthetic process at column 6, line 57 to column 7, line 14 of the patent refers to "reacting a compound of the" second formula given above (emphasis

added) with squaric acid; there is no suggestion that two different compounds of this formula might be used.

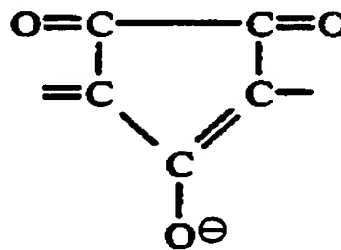
Katagiri describes an optical recording element which uses in its dye layer, *inter alia*, a dye of the formula:



in which A<sub>3</sub><sup>−</sup> represents:



or



Z<sub>93</sub> and Z<sub>94</sub> each represent sulfur, oxygen, or selenium;

Z<sub>91</sub> represents an atomic group necessary to complete a pyrylium, thiapyrylium, selenapyrylium, benzopyrylium, benzothiapyrylium, benzoselenapyrylium, naphthopyrylium, naphthothiapyrylium, or naphthoselenapyrylium ring which may be substituted or not;

Z<sub>92</sub> represents an atomic group necessary to complete a pyrane, thiapyrane, selenapyrane, benzopyrane, benzothiapyrane, benzoselenapyrane, naphthopyrane, naphthothiapyrane, or naphthoselenapyrane ring which may be substituted or not;

R<sub>91</sub>, R<sub>92</sub>, R<sub>93</sub>, and R<sub>94</sub> each represent hydrogen, alkyl, alkoxyl, or a substituted or unsubstituted aryl, styryl, 4-phenyl-1,3-butadienyl, or heterocyclic residue, where R<sub>91</sub> coupled with R<sub>92</sub> and R<sub>93</sub> coupled with R<sub>94</sub> can form each a substituted or unsubstituted benzene ring; and



v and w are each 1 or 2.

Although the aforementioned Formula 9 of Katagiri theoretically covers both symmetric and asymmetric dyes, Columns 75-82 of Katagiri set out the formulae of 22 specific dyes of this formula, and every single one of these dyes is symmetric. Furthermore, the only process for preparing dyes of Formula 9 described in Katagiri is essentially the same as that in Gravesteijn, namely the condensation of squaric (or croconic) acid with two moles of the corresponding heterocyclic salt (see Katagiri, column 81, line 44 to column 83, line 3, and Preparation Examples 4 and 5 in column 83).

## **ISSUES**

The issues to be decided in this appeal are:

(a) Whether the phrase “essentially free from” in claim 16 renders claims 16-23 indefinite for failing to point out and distinctly claim the subject matter which applicants regard as their invention, contrary to 35 USC 112, second paragraph (for convenience, this rejection may hereinafter be called the “112 rejection”); and

(b) Whether claims 15-19 and 24 are unpatentable over Gravesteijn (hereinafter called the “Gravesteijn rejection”); and

(c) Whether claims 15-21 and 23-24 are unpatentable over Katagiri (hereinafter called the “Katagiri rejection”).

It is believed that a full statement of each of these rejections is contained in the following extracts from the Office Actions:

### **(a) The 112 Rejection**

In the first Office Action:

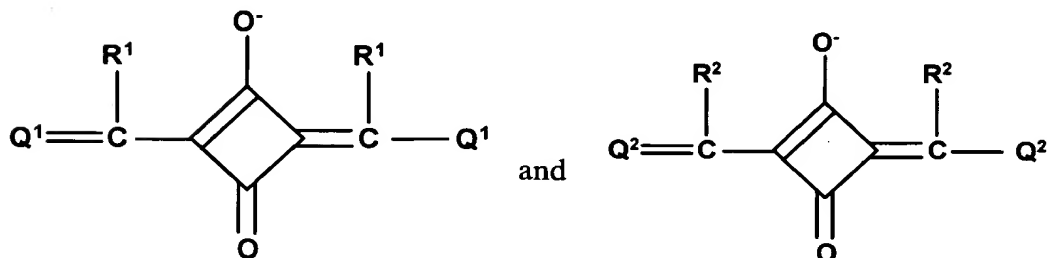
Claims 16-23 are rejected under 35 U.S.C. § 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention. Claim 16 states “A squarylium compound according to claim 15 which is essentially free from squarylium compounds...”, it is unclear what encompasses “essentially free”, if it encompasses a certain purity level or if it encompasses a particular percent conversion of starting materials to final products. Claims which depend from an indefinite

claim are also indefinite. Ex parte Cordova, 10 USPQ2d 1949, 1952 (P.T.O. Bd. App. 1988).

In the final Office Action:

The rejection of claims 16-23 under 35 U.S.C. §112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention stands for the reasons of record and the following.

Applicants argue that in *Marosi*, the Court decided that in the claimed process the Applicants could include the language in the claim "a silicon dioxide source essentially free from alkali metal." The facts of *Marosi* refer to a chemical process wherein the starting materials contain essentially no alkali metal and therefore the final product obtained from the claimed process also contain essentially no alkali metal. Applicants attempt to correlate the fact situation of the present case to that of *Marosi* by stating that they have devised a synthesis which enables asymmetric dyes to be produced essentially without contamination by the corresponding symmetric dyes and hence should be able to claim the resultant, essentially pure dyes which, to the best of their knowledge, has not been and cannot be prepared by the prior art synthesis, which inevitably produces a tertiary mixture. Applicants are claiming the product, i.e., the compounds of formula (I), not a product-by-process. Therefore, the reliance on the *Marosi* case appears to be misplaced. Since the present claims are directed to compounds of formula (I), a person skilled in this art would interpret this claim to include compounds wherein  $Q^1CR^2$  and  $Q^2CR^2$  are different, therefore it would be redundant to state that the compounds of formula (I) are essentially free of compounds of formulae:



In addition, the above formulae read on compounds of the present invention since claim 15 states  $Q^1$  and  $Q^2$  are each independently a pyrylium, thiopyrylium, selenopyrylium, benzpyrylium, benzthiopyrylium or benzselenopyrylium nucleus, and  $R^1$  and  $R^2$  are each

independently a hydrogen or an aliphatic or cycloaliphatic group. One can envision that  $Q^1$  at each occurrence is different or  $Q^2$  at each occurrence is different,  $R^1$  at each occurrence is different, or that  $R^2$  at each occurrence is different, since  $Q^1$ ,  $Q^2$ ,  $R^1$ , and  $R^2$  are independently selected.

(b) and (c) The Gravesteijn and Katagiri Rejections

In the first Office Action:

Claims 15-19 and 24 are rejected under 35 U.S.C. § 103 as being unpatentable over Gravesteijn et al. (US 4,508,811). Gravesteijn et al. disclose alkylpyrylium-squarylium and alkylthiopyrylium-squarylium compounds. Specifically the compounds of formula A of Gravesteijn et al. correspond to Applicants' compounds wherein  $Q^1$  and  $Q^2$  of Applicants' claim 15 are alkyl substituted pyrylium compounds. Applicants require that the  $Q^1CR^1$  and  $Q^2CR^2$  moieties are different, as Gravesteijn et al. disclose that the compounds of formula A have the variable X which is defined as oxygen or sulfur, one can envision that one X of Gravesteijn et al. is oxygen and the other X of Gravesteijn et al. is sulfur. Further, while both of X can be the same, the R groups of Gravesteijn et al. could represent in one instance butyl and in another instance pentyl. It appears that this specification provides for Applicants invention wherein  $Q^1CR^1$  and  $Q^2CR^2$  are different from one another, specifically, wherein the X or variables are different or wherein each occurrence of R is different. One can envision that the compounds of formula A of Gravesteijn et al. would encompass compound (a) of claim 24 wherein R is t-butyl at each occurrence and that one of X is S and the other is O. Therefore, to prepare the instant compounds and expect that they would be useful as dyes would have been prima facie obvious to one of ordinary skill in the art at the time the invention was made since Gravesteijn et al. disclose (col. 2, lines 16-29) the compounds of formula A which are old and known to be useful as dyes.

Claims 15-21 and 23-24 are rejected under 35 U.S.C. § 103 as being unpatentable over Katagiri et al. (US 5,079,127). Katagiri et al. disclose (col. 5, lines 7-53) compounds of formula (9), which generically embrace those as claimed herein. In particular,  $A_3^-$  (of formula (9)) is the moiety as represented at column 5, line 20, and the moieties encompassing  $Z_{91}$  and  $Z_{92}$  are as defined, at column 5, lines 36-46. Specifically,  $Z_{91}$  and  $Z_{92}$  encompass pyrylium, thiopyrylium, selenapyrylium, benzopyrylium, benzothiopyrylium, benzoselenapyrylium, naphthopyrylium, naphthothiopyrylium, naphthoselenapyrylium which bear  $R_{91}$ - $R_{94}$ . Column 5, lines 47-51 define wherein

"each represent hydrogen, alkyl, alkoxy, or a substituted or unsubstituted aryl, styryl, 4-phenyl-1,3-butadienyl, or heterocyclic residue, ...". Applicants claims 20 and 21 require that one of Q<sup>1</sup> or Q<sup>2</sup> contains a phenyl in the 2-position (claim 20) and that the phenyl ring contains an *ortho* alkoxy or cycloalkoxy substituent (claim 21). Since Katagiri et al. define the: each R<sub>91</sub>-R<sub>94</sub> can be a substituted aryl, one can envision an alkoxy substituted phenyl ring on the pyrylium moiety, especially since Katagiri et al. define (col. 74, lines 42-49) substituted aryl to include tolyl, xylyl, biphenyl, ethylphenyl, methoxyphenyl, ethoxyphenyl, amyloxyphenyl, dimethoxyphenyl and others. Therefore, to prepare the instant compounds and expect that they would be useful as dyes in optical recording medium would have been *prima facie* obvious to one of ordinary skill in the art at the time the invention was made since Katagiri et al. teach the substituted pyrylium-squarylium compounds to be old and useful as a dye in an organic thin film used in optical recording medium, particularly the compounds of formula (9), wherein A<sub>3</sub><sup>-</sup> is the squarylium moiety as defined at column 5, line 20 and R<sub>91</sub>-R<sub>94</sub> represent an atomic group which completes a pyrylium, thiapyrylium, selenapyrylium, benzopyrylium, benzothiopyrylium, benzoselenapyrylium, naphthopyrylium, naphthothiopyrylium, naphthoselenapyrylium, which can bear the substituents, e.g., an alkoxy substituted phenyl as defined at column 74, lines 37-59.

Absent a clear explanation of how the instant invention differs from that taught in the prior art and how these differences render the instant invention non-obvious, all claims must be refused.

In the final Office Action:

Claims 15-19 and 24 stand rejected under 35 U.S.C. § 103 as being unpatentable over Gravesteijn et al. (US 4,508,811) for the reasons of record and the following. Claims 15-21 and 23-24 are rejected under 35 U.S.C. § 103 as being unpatentable over Katagiri et al. (US 5,079,127) for the reasons of record and the following. Applicants state that none of the references alone or in combination teach a person having ordinary skill in the relevant art how to prepare the asymmetric dyes of the present invention essentially free from the corresponding symmetrical dyes as required by the present claims. The claims present herein are directed to the squarylium compounds, not a process of preparing the compounds. Further, Applicants admit that the compounds could be prepared by the prior art process yielding a mixture of three products and that the prior art does not disclose how to separate the mixture. Gravesteijn et al. and Katagiri et al. each teach compounds wherein Q<sup>1</sup> and Q<sup>2</sup> of Applicants formula (I) is different. In addition, Applicants have failed to provide evidence that the process of

the prior art or a modified process of the prior art would not produce the asymmetric dyes. Further, since in a patent it is presumed that a process if used by one skilled in the art will produce the product or result described therein, such presumption is not overcome by a mere allegation that it is possible to operate within the disclosure without obtaining the alleged product. It is to be presumed also that skilled workers would as a matter of course, if they do not immediately obtain desired results, make certain experiments and adaptations, within the skill of the competent worker. The failures of experimenters who have no interest in succeeding should not be accorded great weight. **Bullard v. Coe**, 1945 C.D. 13, 64 USPQ 359; **In re Michalek**, 1974 C.D. 458, 74 USPQ 107, 34 CCPA 1124; **In re Reid**, 1950 C.D. 194, 84 USPQ 478, 37 CCPA 884.

## **GROUPING OF CLAIMS**

Pursuant to 37 CFR 1.192(c)(5), applicants submit that, in view of the various rejections set forth above, the patentability of at least each of the following four groups of claims must be assessed separately:

- a) Claims 15 and 24, which are not subject to the 112 rejection, but are subject to the Gravesteyn and Katagiri rejections;
- b) Claim 22, which is subject only to the 112 rejection;
- c) Claim 23, which is subject to the 112 rejection and the Katagiri rejection, but not to the Gravesteyn rejection; and
- d) Claims 16-21, which are subject to all three rejections.

## **ARGUMENT**

### **Summary**

(a) Claim 16 is clear to a person skilled in the art and uses language which the case law holds to be permissible in patent claims.

The claims are patentable over the art of record because (b) the references show that the applicants' statement of the state of the art is correct; (c) neither reference clearly discloses an asymmetric dye in accordance with the present claims, and indeed the references display a studied ambiguity regarding the possibility of producing such asymmetric dyes; (d) neither reference teaches a process for preparing a reasonably pure asymmetric dye; and (e) if one assumes that the references

do disclose the asymmetric dyes, one must also assume that they fail to teach a skilled person how to produce such dyes, and therefore violate the first paragraph of 35 USC 112.

**Relevant case law**

*In re Marosi, Stabenow, and Schwarzmann*, 218 USPQ 289 (CAFC 1983) (hereinafter cited as “*Marosi*”) specifically held the phrase “essentially free of” not indefinite in a chemical claim.

In *Marosi*, the prior art processes for preparing zeolites required the presence of alkali metal in the reaction mixture, and the resulting alkali metal in the zeolite product had to be removed in a separate step before the zeolite was suitable for its intended use as a catalyst (see *Marosi* at 218 USPQ 290, right column). The applicants discovered that by using “a silicon dioxide source essentially free from alkali metal” in the process, it was possible to prepare an essentially alkali metal free product which did not require the alkali metal removal step. The problem applicants faced was that all commercial useful sources of silicon dioxide contained trace amounts of alkali metal, typically of the order of a few parts per million. Accordingly, the applicants defined the invention by the phrase “a silicon dioxide source essentially free from alkali metal”. The evidence showed that the prior art reaction mixtures contained about 3,800 parts per million of sodium, whereas applicants’ mixtures contained about 4 ppm.

The Court allowed the claim containing the phrase “essentially free of alkali metal” despite admitting that it was impossible to draw an exact numerical upper limit on the alkali metal content of the processes falling within the claim, stating:

[A]ppellants have provided a general guideline and examples sufficient to enable a person of ordinary skill in the art to determine whether a process uses a silicon dioxide source “essentially free of alkali metal” to make a reaction mixture “essentially free of alkali metal”. We are persuaded that such a person would draw the line between unavoidable impurities in starting materials and essential ingredients. (*Marosi* at 218 USPQ 292, right column, paragraph denoted “[2]”)

## Detailed argument

### *(a) Claim 16 is clear to a person skilled in the art*

Claim 16 is directed to asymmetric dyes of the formula given in claim 15 essentially free from the corresponding symmetrical dyes of the two formulae given in claim 16. Contrary to the Examiner's argument quoted above, it is not redundant to state that the compounds of Formula (I) are essentially free from symmetrical dyes of the two formulae given in claim 16. A claim to a chemical compound normally extends to that compound in a mixture; for example, clearly claim 15 would be infringed by anyone making a solution of one of the asymmetric dyes in (say) acetone. Similarly, claim 15 would be infringed by anyone producing one of the asymmetric dyes containing as an impurity (say) 1 percent by weight of one of the corresponding symmetric dyes.

In some applications of infra-red dyes, the presence of even minor amounts of symmetric by-products in the desired asymmetric dye may cause significant problems. For example, as already noted, in the thermal imaging media described in U.S. Patents Nos. 4,602,263 and 4,826,976, three separate imaging layers are present, having infra red absorbers with absorptions at 760, 820 and 880 nm. Conveniently, two of these three absorbers are Dye A shown in Figure 1 of this application, in which  $R^1$  and  $R^2$  are each a hydrogen atom, and the corresponding bis selenopyrylium dye. However, if Dye A is contaminated with even a small proportion of one of the symmetrical bis selenopyrylium dyes, serious problems may result in such a medium, in that the symmetrical bis selenopyrylium impurity in the layer containing Dye A will absorb the "wrong" radiation, which may lead to unwanted exposure of parts of the layer containing Dye A and a reduction in sensitivity of the medium because the bis selenopyrylium impurity will absorb a large part of the radiation intended to cause color change in a different color forming layer.

As discussed in more detail below with reference to the 35 USC 103 rejections, the applicants have devised a synthesis which enables the asymmetric dyes of claim 15 to be produced essentially without contamination by the corresponding

symmetric dyes, and hence desire to claim the resultant, essentially pure dye. Although, given the ability of modern analytical techniques (for example gas chromatography-mass spectrometry) to detect impurities down to the parts per trillion level, the applicants cannot guarantee that all dyes produced by their new process are absolutely free from the corresponding symmetric dyes (which could conceivably be produced by minor side reactions), they have found that the dyes they produce are essentially pure, and can be used in all applications of such dyes without interference from the symmetrical dyes. In these circumstances, although it may not be possible to specify an exact numerical limit for “essentially free from”, the gap between the claimed dyes and the prior art mixtures, which cannot contain more than about fifty percent of the desired asymmetric dye, is so great that *Marosi* applies, and the phrase “essentially free from” is not indefinite in the context in which it is used.

The Examiner’s attempt to distinguish the present case from *Marosi* on the ground that *Marosi* claimed a process whereas the present claim claims a composition is illogical. In *Marosi*, the phrase was applied to the starting material used in a process, not to a process condition, and hence the phrase was used to define the nature of a chemical composition. There is no reason why the same phrase should be permissible to define the nature of a starting material in a process, yet impermissible to define the nature of a composition claimed *per se*.

Finally, the Examiner’s statement that “One can envision that  $Q^1$  at each occurrence is different or  $Q^2$  at each occurrence is different,  $R^1$  at each occurrence is different, or that  $R^2$  at each occurrence is different, since  $Q^1$ ,  $Q^2$ ,  $R^1$ , and  $R^2$  are independently selected”. While it is common practice to have a general formula containing multiple occurrences of a variable (say  $R$ ) and to define the formula by wording such as “each group  $R$  is independently selected from . . .”, there is no rule of claim construction which requires one to imply the word “independently” where the claim does not contain this word. Indeed, the natural construction would be the other way; where the claim does not contain the word “independently” it should be presumed that all occurrences of the same variable represent the same group. In the



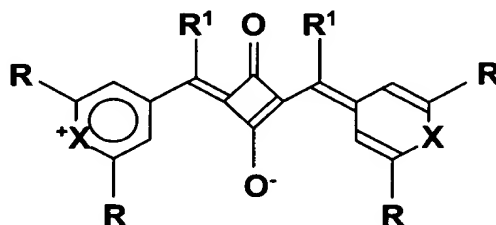
present case, it is apparent from the language of claims 15 and 16, and the corresponding portion of the description at page 30, lines 1-8, that the formulae in claim 16 represent only symmetrical dyes, in which the two groups  $Q^1$  are the same, the two groups  $Q^2$  are the same, the two groups  $R^1$  are the same, and the two groups  $R^2$  are the same. This is shown by the explicit statements in claim 15 that  $Q^1$  and  $Q^2$  are independently selected, and that  $R^1$  and  $R^2$  are independently selected, and the absence from claim 16 of any language implying such independent selection. Also, the aforementioned passage on page 30, which is manifestly equivalent to claim 16, states that "Because the present dye-forming reaction does not produce the compounds of Formula I admixed with the corresponding symmetric compounds, the present dye-forming reaction can readily produce compounds of Formula I essentially free from squarylium compounds of the formulae" given in claim 16, thereby showing that the formulae in claim 16 represent symmetric dyes.

***(b) The references show that the applicants' statement of the state of the art is correct***

The two references both show that the applicants' description of the state of the art in the introductory part of this application (essentially reproduced above) is correct; in each case, the only method described for the synthesis of the dyes is condensation of two moles of the corresponding 4-methylpyrylium (etc.) salt with one mole of squaric acid.

***(c) Neither reference clearly discloses an asymmetric dye***

As noted above, Gravesteijn describes an optical recording element (i.e., a compact disc) which uses in its dye layer a squarylium dye of formula:



in which R is an alkyl group having at least three carbon atoms, and  $R_1$  is a hydrogen atom or methyl. In this formula, "X is an oxygen atom or a sulfur atom" (see column

2, line 26 and claim 1 of Gravesteijn), but nowhere in the patent is there any language which resolves the ambiguity as to whether the two atoms X must be the same or can be different. The Examples show only preparations of dyes in which both atoms X are sulfur (Example 1) or oxygen (Example 2). Furthermore, the detailed description of the synthetic process at column 6, line 57 to column 7, line 14 of the patent refers to "reacting *a* compound of the" (emphasis added) formula for the pyrylium salt with squaric acid, which implies that the process is only intended to be carried with a single pyrylium salt, and hence will produce a symmetric dye. Certainly, nowhere in Gravesteijn is there any discussion of the problems of separating the ternary mixture which will inevitably result if this process were carried out using a mixture of two salts.

An exactly parallel situation exists with regard to Katagiri.  $Z_{93}$  and  $Z_{94}$  have the same definition,  $Z_{91}$  and  $Z_{92}$  have the same definition (the difference between, for example "pyrylium" in the definition of  $Z_{91}$  and "pyrane" in the definition of  $Z_{92}$  is of course illusory; all the relevant compounds are resonance hybrids and the positive charge is distributed equally between the two pyrylium residues), and  $R_{91}$ ,  $R_{92}$ ,  $R_{93}$  and  $R_{94}$ , all have the same definition, and nowhere in the patent is there any language which resolves the ambiguity as to whether the two similar groups must be the same or can be different. However, although Formula 9 of Katagiri theoretically covers both symmetric and asymmetric dyes, the 22 specific dyes of this formula set out in columns 75-82 are, without exception, symmetric. Furthermore, the only process described for preparing these dyes is essentially the same as that in Gravesteijn, namely the condensation of squaric (or croconic) acid with two moles of the corresponding heterocyclic salt. Note also that the same ambiguous situation exists with regard to the croconate dyes of Formulae 1 and 2 of Katagiri, which are prepared by an analogous process; all 34 dyes in columns 8-18 are symmetric.

Both Gravesteijn and Katagiri are assigned to multinational companies (Gravesteijn to U.S. Philips Corporation, the North American subsidiary of Philips

Gloelampenfabriken N.V. of the Netherlands, and Katagiri to Canon Kabushiki Kaisha) and appear to be products of sophisticated research teams. To find one such reference which displays an odd ambiguity about whether the reference covers asymmetric dyes would be unusual; to find two such references strains credibility. It is difficult to avoid the conclusion that the ambiguity regarding asymmetric dyes in Gravesteijn and Katagiri is a deliberate, studied ambiguity; the inventors were no doubt aware of the possibility of producing asymmetric dyes, but were equally aware of their failure to provide a reasonably synthesis of such dyes, and deliberately fudged the issue.

***(d) Neither reference teaches a process for preparing a reasonably pure asymmetric dye***

As already noted, the only process described in the references for producing the relevant dyes is condensation of one mole of squaric acid with two moles of the appropriate pyrylium (etc.) salt, and it should be obvious to any chemist that, as stated in the introduction to this application, if one tries to synthesize an asymmetric dye in this manner, one will obtain a mixture of three different but similar products (namely the desired asymmetric dye and the two symmetric dyes) which will be very difficult to separate; isolation of the asymmetric dye will especially pose a problem in that all of properties would be expected to be intermediate those of the two symmetric dyes. With respect, the Examiner's statement that "Applicants have failed to provide evidence that the process of the prior art or a modified process of the prior art would not produce the asymmetric dyes" ignores this obvious problem. No reasonable chemist would attempt to produce a mixed ester of a dicarboxylic acid (say ethyl butyl oxalate) by reacting oxalic acid with a mixture of ethanol and butanol because separation of the desired ethyl butyl oxalate from diethyl oxalate and dibutyl oxalate would pose extraordinary difficulties.

This is not a case where the art clearly teaches that a particular reaction can be carried out and applicants dispute this. As noted above, there is no unambiguous teaching in the references regarding the preparation of asymmetric dyes, and certainly no worked example of such a preparation. Accordingly, the three cases

cited by the Examiner are not analogous to the present situation. In *Bullard v. Coe*, 64 USPQ 359 (D.C. Ct. App. 1945), it was alleged that a mechanical invention shown in a prior patent was unworkable, although there was no obvious reason on the face of the document why the device should not work. In *In re Michalek*, 74 USPQ 107 (CCPA 1947), the reference clearly stated that a particular dichlorostyrene was polymerizable, and applicants maintained that it was not. In *In re Reid*, 84 USPQ 478 (CCPA 1950), employees of the assignees filed affidavits alleging that the disclosures of various references produced inoperative or unsatisfactory results. In the present case, the applicants are not suggesting that the process described in the references does not, in general, work, and they in no way suggest that the patentees did not produce the specific compounds stated to have been produced. Applicants do, however, maintain that, for reasons which should be clear to any experienced chemist, when the process of the references is applied to a sub-group of compounds, namely asymmetric dyes, an unworkable mixture of products results, and that the references give no hint as to how to deal with this problem. The applicants are not aware of any reported case which is squarely on point in this situation.

***(e) If the references do disclose asymmetric dyes, they violate 35 USC 112***

If, as the Examiner maintains but contrary to the applicants' arguments above, the two references do disclose asymmetric dyes, but teach only a process which, when applied to the synthesis of such asymmetric dyes, yields only an unworkable mixture, the references are invalid for failure to comply with the first paragraph of 35 USC 112, in that they fail to enable a person skilled in the art to make and use the invention, namely a useable form of the asymmetric dye. This appears an unnatural interpretation of the references, given that issued patents are presumed valid (35 USC 282).

## **CONCLUSION**

For all of the foregoing reasons, the rejections of the claims on appeal should be reversed and the application allowed.

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Respectfully submitted,

A handwritten signature in cursive script, appearing to read "David J. Cole".

David J. Cole  
Registration No. 29629

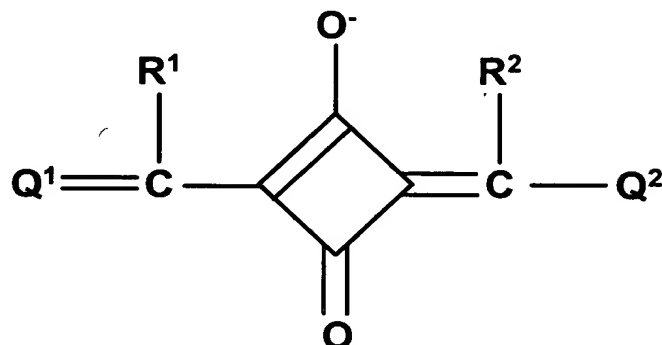
Polaroid Corporation  
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## APPENDIX

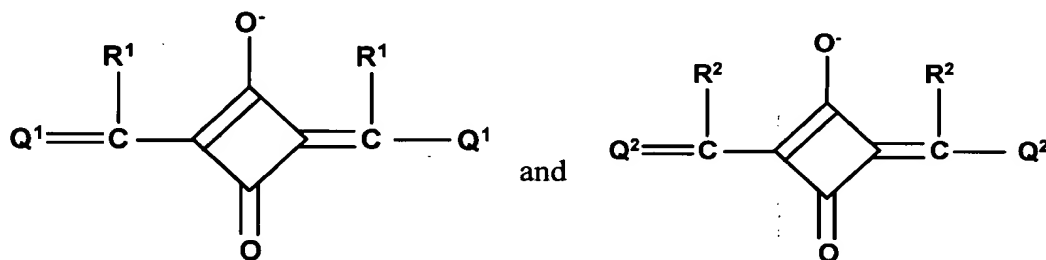
### Claims on Appeal

- 1                    15.    A squarylium compound of the formula:



2  
3    wherein Q¹ and Q² are each independently a pyrylium, thiopyrylium, selenopyrylium,  
4    benzpyrylium, benzthiopyrylium or benzselenopyrylium nucleus, and R¹ and R² are  
5    each independently a hydrogen atom or an aliphatic or cycloaliphatic group, the  
6    Q¹CR¹ grouping being different from the Q²CR² grouping.

- 1                    16.    A squarylium compound according to claim 15 which is  
2    essentially free from squarylium compounds of the formulae:



- 3  
1                    17.    A squarylium compound according to claim 16 wherein each of  
2    Q¹ and Q² is a 4-pyrylium, 4-thiopyrylium, 4-selenopyrylium, 4-benzpyrylium,  
3    4-benzthiopyrylium or 4-benzselenopyrylium nucleus.
- 1                    18.    A squarylium compound according to claim 17 wherein at least  
2    one of Q¹ and Q² is a 2,6-dialkylpyrylium, -thiopyrylium or -selenopyrylium nucleus,  
3    in which each of the alkyl groups contains not more than about 8 carbon atoms.

1 19. A squarylium compound according to claim 18 wherein at least  
2 one of Q<sup>1</sup> and Q<sup>2</sup> is a 2,6-di-tertiary butylpyrylium, -thiopyrylium or -selenopyrylium  
3 nucleus.

1 20. A squarylium compound according to claim 17 wherein one of  
2 Q<sup>1</sup> and Q<sup>2</sup> is a 2-phenyl benzpyrylium, benzthiopyrylium or benzselenopyrylium  
3 nucleus and the other is (a) a 2-substituted benzpyrylium, benzthiopyrylium or  
4 benzselenopyrylium nucleus, in which the 2-substituent is an alkyl, alkenyl, alkynyl or  
5 alkicyclic group, or (b) a 2,6-dialkyl butylpyrylium, -thiopyrylium or -selenopyrylium  
6 nucleus.

1 21. A squarylium compound according to claim 20 wherein the 2-  
2 phenyl group has an *ortho* alkoxy or cycloalkoxy substituent.

1 22. A squarylium compound according to claim 17 wherein one of  
2 Q<sup>1</sup> and Q<sup>2</sup> is a benzpyrylium, benzthiopyrylium or benzselenopyrylium nucleus  
3 bearing at its 7-position an -N[(CH<sub>2</sub>)<sub>3</sub>]<sub>2</sub> grouping in which the ends of the  
4 trimethylene groups remote from the nitrogen atom are joined to the 6- and  
5 8-positions of the nucleus, so that the -N[-(CH<sub>2</sub>)<sub>3</sub>]<sub>2</sub> grouping and the phenyl ring of  
6 the nucleus together form a julolidine ring system, and the other is (a) a 2-substituted  
7 benzpyrylium, benzthiopyrylium or benzselenopyrylium nucleus, in which the 2-  
8 substituent is an alkyl, alkenyl, alkynyl or alicyclic group, or (b) a 2,6-dialkyl  
9 butylpyrylium, -thiopyrylium or -selenopyrylium nucleus.

1 23. A squarylium compound according to claim 17 wherein at least  
2 one of Q<sup>1</sup> and Q<sup>2</sup> is a benzpyrylium, benzthiopyrylium or benzselenopyrylium nucleus  
3 bearing at its 6-position an alkoxy or cycloalkoxy group.

1 24. A squarylium compound according to claim 15 in which:

2 (a) Q<sup>1</sup> is a 2,6-bis(1,1-dimethylethyl)-4-pyrylidene grouping, Q<sup>2</sup> is  
3 a 2,6-bis(1,1-dimethylethyl)-4-thiopyrylium grouping, and R<sup>1</sup> and R<sup>2</sup> are each a  
4 hydrogen atom, namely [4-[[3-2,6-bis(1,1-dimethylethyl)-(4H-pyran-4-ylidene)-  
5 methyl]-2-hydroxy-4-oxo-2-cyclobuten-1-ylidene]methyl]-2,6-bis(1,1-dimethyl-  
6 ethyl)thiopyrylium hydroxide inner salt;

7 (b)  $Q^1$  is a 2,6-bis(1,1-dimethylethyl)-4-pyrylidene grouping,  $Q^2$  is  
8 a 2,6-bis(1,1-dimethylethyl)-4-selenopyrylium grouping, and  $R^1$  and  $R^2$  are each a  
9 hydrogen atom, namely 4-[[3-2,6-bis(1,1-dimethylethyl)-(4H-pyran-4-ylidene)-  
10 methyl]-2-hydroxy-4-oxo-2-cyclobuten-1-ylidene]methyl]-2,6-bis(1,1-dimethylethyl)-  
11 selenopyrylium hydroxide inner salt;

12 (c)  $Q^1$  is a 7-diethylamino-2-(1,1-dimethylethyl)benz[b]-4H-pyran-  
13 4-ylidene grouping,  $Q^2$  is a 7-diethylamino-2-phenylbenzpyrylium grouping, and  $R^1$   
14 and  $R^2$  are each a hydrogen atom, namely 4-[3-[[7-diethylamino-2-(1,1-dimethyl-  
15 ethyl)benz[b]-4H-pyran-4-ylidene]methyl]-2-hydroxy-4-oxo-2-cyclobuten-1-ylidene]-  
16 methyl]7-diethylamino-2-phenylbenzpyrylium hydroxide inner salt dye;

17 (d)  $Q^1$  is a 2,6-bis[1,1-dimethylethyl]-4-selenopyrylidene grouping,  
18  $Q^2$  is a 2-[2-trifluoromethylphenyl]benz[b]pyrylium grouping, and  $R^1$  and  $R^2$  are each  
19 a hydrogen atom, namely 4-[[3-[2,6-bis[1,1-dimethylethyl]-[4H-selenopyran-  
20 4-ylidene]methyl]-2-hydroxy-4-oxo-2-cyclobuten-1-ylidene]methyl]-2-[2-  
21 trifluoromethylphenyl]benz[b]pyrylium hydroxide inner salt dye;

22 (e)  $Q^1$  is a 6-[but-2-oxy]-2-[1,1-dimethylethyl]benz[b]-4H-pyran-4-  
23 ylidene grouping,  $Q^2$  is a 6-[2-ethylbut-1-oxy]-2-phenylbenzpyrylium grouping, and  
24  $R^1$  and  $R^2$  are each a hydrogen atom, namely 4-[[3-[[6-[but-2-oxy]-2-[1,1-  
25 dimethylethyl]benz[b]-4H-pyran-4-ylidene]methyl]-2-hydroxy-4-oxo-2-cyclobuten-  
26 1-ylidene]methyl]-6-[2-ethylbut-1-oxy]-2-phenylbenzpyrylium hydroxide inner salt  
27 dye; and

28 (f)  $Q^1$  is a 2,6-bis[1,1-dimethylethyl]-4-thiopyrylidene grouping,  
29  $Q^2$  is a 2,6-bis[2,4-dimethylphenyl]pyrylium grouping, and  $R^1$  and  $R^2$  are each a  
30 hydrogen atom, namely 4-[[3-[2,6-bis[1,1-dimethylethyl]-[4H-thiopyran-4-ylidene]-  
31 methyl]-2-hydroxy-4-oxo-2-cyclobuten-1-ylidene]methyl]-2,6-bis[2,4-  
32 dimethylphenyl]pyrylium hydroxide inner salt dye.



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**CERTIFICATE OF MAILING**

I hereby certify that this paper, dated June 13, 1997, is being deposited with the United States Postal Service as first class mail in an envelope addressed to Assistant Commissioner for Patents, Washington DC 20231, on *June 13, 1997*.

*David J. Cole*

David J. Cole